Neutron reflectivity on $CoFe_2O_4$ exchange springs for spin valve applications

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Insulating $CoFe_2O_4$ is a candidate for biasing ferromagnetic layers in spin valves because it does not shunt current. Ferrimagnetic $CoFe_2O_4$ pins the neighboring ferromagnetic layer via an exchange-spring mechanism. We have examined the field-dependent layer switching in a $CoFe_2O_4/CoFe_{10}$ bilayer using back/front polarized neutron reflectometry. A magnetic twist is evident in intermediate fields and gradually collapses as the field is increased. However, the twist is confined mostly to the interface or to the magnetically hard $CoFe_2O_4$ layer. This result contrasts with the expectation that the magnetic twist should reside in the soft layer in the field region of magnetic reversibility. © *2004 American Institute of Physics.* [DOI: 10.1063/1.1669127]

The discovery of giant magnetoresistance (GMR) motivated the development of spin valves for applications such as magnetic recording read heads. A simple spin valve usually consists of a biasing layer, a pinned ferromagnetic layer, a conductive nonmagnetic spacer layer, and a freely rotating ferromagnetic layer. While most spin valves rely upon a metallic antiferromagnetic biasing layer, such as PtMn, use of an insulating biasing layer could lead to potentially higher GMR values for spin valves with a current in-plane geometry, since it does not shunt current. A promising candidate is hard ferrimagnetic cobalt ferrite ($CoFe_2O_4$), which couples to the neighboring, soft ferromagnetic layer to form an exchange-spring bilayer. A prototypical exchange spring[1] is composed of a hard magnet with a large coercive field that is exchange coupled to a soft magnet with a high saturation moment. According to a model advanced by Kneller and Hawig,[1] in a modestly applied reverse field the magnetization of the soft material should twist away from that of the hard material. Since theory suggests that the twist is confined to the soft layer in this field region, the magnetization of the soft layer should be reversible. If the field is thus cycled over a limited range, the hard layer should effectively pin the soft layer since the twist is confined primarily to the soft layer and the magnetization of the hard layer remains in the direction of the initial field.

To demonstrate the feasibility of this pinning mechanism, we recently grew a $CoFe_2O_4/Co/Cu/Co/Ni_{80}Fe_{20}$ spin valve[2,3] that shows high GMR, excellent biasing, and good free-layer properties. Optimization of this device requires examination of the field-dependent switching of the individual magnetic layers and characterization of the resultant magnetic twist. Since bulk magnetization measurements are sensitive only to the net magnetization of the entire sample, information pertaining to the depth dependence of the magnetic twist and its chirality have been obtained from other techniques[4,5] including magneto-optical indicator film imaging.[6,7] As demonstrated for an $Fe_{55}Pt_{45}/Ni_{80}Fe_{20}$ bilayer,[8] polarized neutron reflectometry (PNR) is particularly well suited for the study of exchange springs and related materials with noncollinear magnetism because the direction and magnitude of the magnetic moment in each layer can be determined as a function of depth with subnanometer resolution. In this study, we examined a $CoFe_2O_4/CoFe_{10}$ bilayer with front/back PNR techniques[8] in order to highlight the pinning process. Our measurements confirm that the moments in the hard ferrimagnetic $CoFe_2O_4$ layer and the soft ferromagnetic $CoFe_{20}$ layer twist smoothly toward the applied field and eventually collapse as the field is varied from 0 to 900 mT. The magnetic twist, however, is confined mostly to the interface or to the hard $CoFe_2O_4$ layer, rather than to the soft $CoFe_{10}$, in fields near and above 50 mT where the magnetization is expected to be reversible. This surprising result contrasts with expectations for a classical exchange spring[1] and modifies our understanding of the pinning process in related oxide-based spin valves.[2,3]

The samples for our PNR studies were grown at Hitachi by reactive dc magnetron sputtering from metallic targets as described elsewhere.[2,3] We considered a series of $CoFe_2O_4(37.5\ \mathrm{nm})/CoFe_{10}(x)/Ta(10.0\ \mathrm{nm})$ exchange-coupled films deposited on Si(100) wafers. In this work we shall focus on the sample with $x = 6.0$ nm. (The layer thick-

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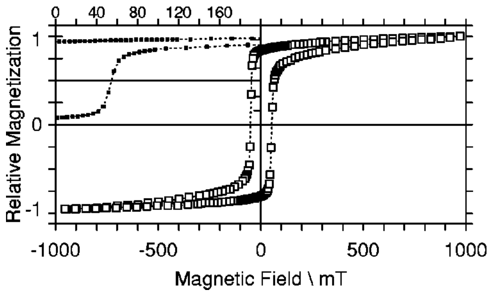

FIG. 1. Magnetic hysteresis of $CoFe_2O_4$(37.5 nm)/$CoFe_{10}$(6.0 nm)/Ta(10.0 nm) measured with VSM. (Inset) The region between 0 and 200 mT.

nesses quoted are nominal.) The bulk magnetic properties of this film parallel to the applied magnetic field were determined at room temperature with vibrating-sample magnetometry VSM. The hysteresis loop is plotted in Fig. 1. The maximum field applied was 1000 mT. Although the sample has clearly not achieved its saturation magnetization, this minor loop accurately reproduces the magnetic field conditions we can apply during the measurement of the PNR. Instrumental conditions there limit the maximum field we can apply to 900 mT. From the data for Fig. 1 we extract a coercive field $H_c = 52.1$ mT.

To verify an individual layer's contribution to the hysteresis curve, PNR was measured at room temperature. We used neutrons of wavelength 0.475 nm with the NG-1 reflectometer at the NIST Center for Neutron Research. The techniques used to polarize the neutrons are described elsewhere.[9] The neutrons are polarized along the vertical direction (i.e., parallel to the axis designated x) in the sample plane, and the data are corrected for the efficiencies of the polarizing elements,[9] which range from 95% to 100%, as well as for the footprint of the beam. The reflectivities R^{++} and R^{--}, in which the polarized neutron does not change its orientation, sense the chemical structure of the film and the x component of the magnetization **M**; they are designated as nonspinflip (NSF) scattering. The reflectivities R^{+-} and R^{-+} are nonzero only when components of **M** lie perpendicular to the x axis (i.e., parallel to the y axis) in the sample plane, and are not particularly sensitive to the chemical structure; they are designated as spinflip (SF) scattering.

To help establish whether there was a smooth twist in magnetization from the ferromagnetic $CoFe_{10}$ through the ferrimagnetic $CoFe_2O_4$, reflectivity from the front and back surfaces were measured in the same field state, as described in Ref. 8. This approach is particularly useful in our case because the small net magnetization in the ferrimagnet produces little magnetic contribution to the reflectivity when compared to the ferromagnet's contribution. The sample was brought to a high-field state of -900 mT before applying fields of -697, 1.1, 29, 47, 54, 70, 100, and 200 mT. The state at -697 mT served to establish the maximum magnetization in each respective layer. This field range is expanded in the inset of Fig. 1.

Figure 2 shows the reflectivity measured at 47 mT, just before the magnetization reverses, and is typical of the data we measured. The reflectivity from the back surface is plotted on the left with scattering vector q increasing towards the left. The reflectivity from the front surface is plotted on the right with q increasing towards the right. Here ''front'' indicates the neutrons encounter the ferromagnetic layer before the ferrimagnetic layer while ''back'' indicates the opposite. The NSF cross sections R^{++} and R^{--} are plotted against the left axis. The SF cross sections R^{+-} and R^{-+} are plotted against the right axis, which has been shifted relative to the NSF axis. For clarity, the uncertainty introduced by the counting statistics has been omitted. Above $q = 1\ \mathrm{nm}^{-1}$ the uncertainty in the SF scattering is equal to the plotted SF reflectivity. As discussed in Ref. 8, differences between the front and back reflectivity indicate canted or twisted magnetic layers. The front SF reflectivity shows a strong peak just at the critical $q \approx 0.19\ \mathrm{nm}^{-1}$, but this peak is absent in the back reflectivity. Experience suggests the peak in this location corresponds to the soft ferromagnetic layer being twisted or canted relative to the field, while the hard ferrimagnetic layer is mainly (anti)parallel to the field.

The reflectivities for all positive fields were fit to a model described in Ref. 8. Salient features include the possibility for a smooth twist in either magnetic layer, constant pitch of the twist (separately variable for each layer), conti-

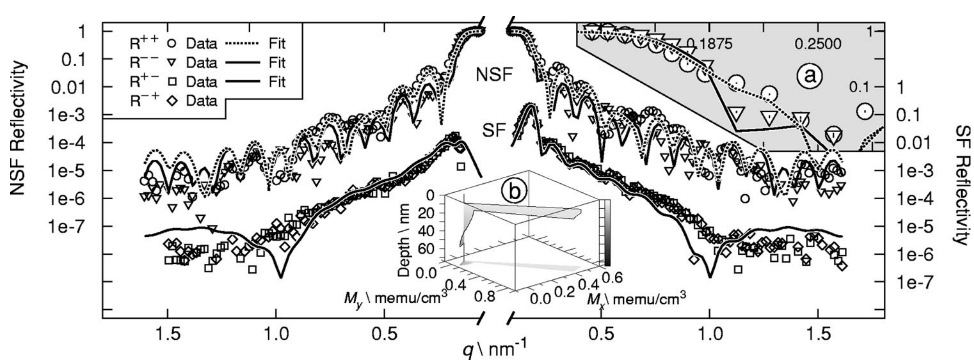

FIG. 2. Reflectivity at 47 mT. Data from the back (front) surface are shown on the left (right) with q increasing towards the left (right). The NSF(SF) data are plotted against the left (right) axes. Note the relative shift of these two axes. Data are plotted with symbols, the fits with lines. (Inset a). Focus near the front NSF critical q which emphasizes the crossing cross sections. (Inset b) Fitted vector magnetization. The lower curve is a projection of the magnetization into the xy sample plane.

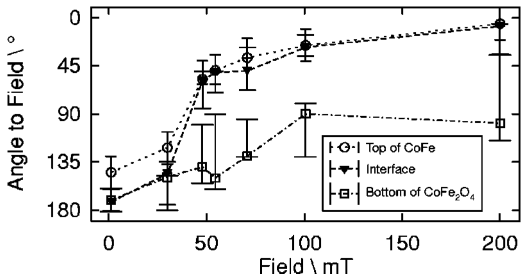

FIG. 3. The angle of the magnetization at the top of the $CoFe_{10}$, the interface, and at the bottom of the $CoFe_2O_4$.

nuity of angle across the interface between the layers, and an untwisted region in each layer at the boundaries with the substrate and Ta layer. All four cross sections from both sides were fit (i.e., lines in Fig. 2) simultaneously by minimizing χ^2. Structural parameters once refined at -697 mT and again at 1.1 mT were thereafter kept fixed for the remaining fields, as was the magnitude of the magnetization. Our fitting models have not allowed for the possibility of a multidomain sample, and it is certainly reasonable to expect the presence of domains at intermediate fields. A further investigation of this possibility is underway. A key feature driving the fits is the crossing of the NSF reflectivities just above the critical q, as shown in inset (a) of Fig. 2. The behavior of the SF reflectivity in this same q range is also an important contributor to the final fitted parameters.

The vector magnetization at 47 mT is plotted in inset (b) of Fig. 2. Two curves are shown. First is the vector magnetization plotted as a function of depth into the sample where gray shading indicates depth. The air–Ta interface occurs at $z=0$ nm. As an aid to following the turning of the magnetization, the projection of the magnetization into the xy sample plane is also shown. We find that a portion of the ferrimagnet near the $CoFe_2O_4/CoFe_{10}$ interface at $z \approx 16$ nm is twisted, while the remainder is untwisted. The fit for this field, however, is not particularly sensitive to whether the small twist in the ferromagnet is concentrated at the interface or dispersed throughout, as depicted in the inset.

For all the fields considered, the fit is most responsive to variations in the following parameters: the angle of the magnetization at the top of the ferromagnet, the interface between ferro- and ferrimagnet, and at the bottom of the ferrimagnet. The best-fit values of these parameters are plotted in Fig. 3 as a function of field. The uncertainty in these parameters was determined from a visual inspection of the fit as the parameters were independently swept through the range of physically reasonable values. For example, the angle of the bottom of the hard ferrimagnet was swept from 180° (i.e., antiparallel to the external field) to the best-fit interfacial value. Similiarly, the angle at the top of the soft ferromagnet was swept from 0° (i.e., parallel to the external field) to the interfacial value. The interfacial angle was varied between the best fit values for the top and bottom layers. The uncertainty in the angle at the bottom of the ferrimagnet is somewhat large. One cause is that the ferrimagnet contributes 1% to the magnetic scattering since its net moment is 1/10 that of the ferromagnet. In addition, two features of the measured data compete in determining the best-fit value of this angle. On the one hand, the smooth nature of the SF scattering, devoid of any oscillations found in the NSF scattering, drives the fit to values close to 180°. On the other hand, the crossing of the NSF cross sections just above the critical q drives the angle towards 90° or 0°. Despite the uncertainty, we conclude that a twist is evident in the hard $CoFe_2O_4$ ferrimagnet in the field region near and above 50 mT.

Figure 3 clearly indicates that below 50 mT a magnetic twist with an angular extent of $<45°$ forms in the soft $CoFe_{10}$. Above 50 mT the top of the ferromagnet gradually approaches full alignment with the external field. The thin nature of this layer makes it difficult to extract whether the interfacial angle is significantly different from the angle in the ferromagnet. This uncertainty may be due to the presence of a multidomain state. It is clear, however, that the twist through the interface and in the ferromagnet collapses almost completely between 75 and 100 mT.

The models of exchange-spring magnets[1] should apply to these two exchange-coupled layers. Those models suggest that in the field region where the magnetization is fully reversible, any twist would most likely occur in the soft magnetic layer. Recent experimental determinations[8] and simulations[10,11] demonstrate a greater participation than predicted by the hard layer in other exchange-spring systems. For our sample, polarized neutron reflectivity studies indicate that the twist, once initiated in the soft $CoFe_{10}$ layer near the coercive field, quickly propagates through it and is then confined to the hard $CoFe_2O_4$ layer. The twist is only slowly driven out of the hard layer as the sample approaches saturation. Understanding of this unexpected behavior requires a detailed theoretical analysis of the competing energetics for the two magnetic layers[11,12] and may provide additional insight into the utilization of these materials as pinning layers.

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